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## Exchange Bias in Fe<sub>0.6</sub>Zn<sub>0.4</sub>F<sub>2</sub> Heterostructures

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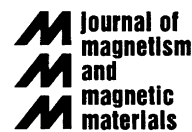
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# Exchange bias in $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2/\text{Fe}$ heterostructures

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## Abstract

The exchange bias field,  $H_e$ , is measured in  $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2/\text{Fe}$  heterosystems prepared from Fe layers of 14 and 5 nm thickness which are deposited on top of the compensated (1 1 0) surface of the antiferromagnet. Deviations from a linear dependence of  $H_e$  on the magnetization of the Fe layer are attributed to ferromagnetic domains. Moreover, piezomagnetism and its influence on  $H_e$  are evidenced. © 2002 Elsevier Science B.V. All rights reserved.

**Keywords:** Exchange biasing; Antiferromagnetism; Ising model; Magnetization – stress induced; Domains

The exchange bias effect is one of the most challenging topics in modern thin film magnetism. It describes a coupling phenomenon between ferromagnetic (FM) and antiferromagnetic (AF) materials which is phenomenologically characterized by a shift  $H_e$  of the ferromagnetic hysteresis loop along the magnetic field axis [1,2]. The exchange bias field  $H_e$  reflects an unidirectional anisotropy which originates from the interface coupling of the ferromagnet and its AF pinning layer. Single crystals covered with metallic FM films are among the favored models of such heterosystems. E.g., Fe and CoPt layers deposited on top of surfaces of  $\text{FeF}_2$  single crystals have been extensively studied [3–5].

The Meiklejohn-Bean (MB) model is a very useful first approach in order to interpret the experimental results [1,2]. In particular, the basic formula  $\mu_0 H_e = -JS_{\text{AF}}S_{\text{FM}}/(M_{\text{FM}}t_{\text{FM}})$  points out the necessity of net magnetic moments  $S_{\text{AF}}$  and  $S_{\text{FM}}$  at the interface on the AF and the FM side as well as the dependence on the FM layer thickness  $t_{\text{FM}}$ , the saturation magnetization  $M_{\text{FM}}$  of the FM layer and the interface coupling  $J$ , respectively. The above expression can be generalized in order to describe the dependence of  $H_e$  on the AF layer thickness and a possible magnetic moment in the AF bulk [6–8]. Recently, it has been stressed, that the surplus magnetic moment of random field domains gives rise to exchange bias in heterostructures with compen-

sated AF interfaces [9–11]. Stimulated from these findings, we investigate the exchange bias effect by Superconducting Quantum Interference Device (SQUID, Quantum Design MPMS-5S) magnetometry in  $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2(1\ 1\ 0)/\text{Fe}5\text{ nm}/\text{Ag}35\text{ nm}$  and in  $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2(1\ 1\ 0)/\text{Fe}14\text{ nm}/\text{Ag}35\text{ nm}$  heterostructures as a function of the temperature and of the magnetic moment,  $m_{\text{FM}}$ , of the ferromagnet.

The samples are grown by UHV-deposition of 5 and 14 nm Fe on top of the compensated (1 1 0) surface of the diamagnetically diluted antiferromagnet which is thermally stabilized at  $T = 425\text{ K}$  during the growth process. After field cooling to below the Néel temperature of  $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2$ ,  $T_N = 47\text{ K}$ , the exchange bias effect shifts the hysteresis loop of the FM film by the amount  $H_e$  along the magnetic field axis. The sign of the shift is determined by the direction of the magnetic moment of the Fe layer,  $m_{\text{FM}}$  [12]. In order to determine  $m_{\text{FM}}$  from measurements of the total moment, the magnetic hysteresis is measured at  $T = 100\text{ K} \approx 2T_N$ . Subsequently, a magnetic field is applied in the non-overshoot mode of the magnetometer in order to follow unambiguously the upper branch of the hysteresis loop from the saturation value  $m_s$  to the target value  $m_{\text{FM}}$  where  $-m_s \leq m_{\text{FM}} \leq m_s = 9.0 \times 10^{-7}\text{ Am}^2$ . The magnetic field that prepares  $m_{\text{FM}}$  is applied during the freezing process.

Fig. 1 exhibits the  $H_e$  vs.  $m_{\text{FM}}$  dependence of the  $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2(1\ 1\ 0)/\text{Fe}14\text{ nm}/\text{Ag}35\text{ nm}$  heterostructure. The data are obtained from the shifts of the magnetic hysteresis curves after cooling the system in the applied field to  $T = 10\text{ K}$ . The inset shows a typical hysteresis

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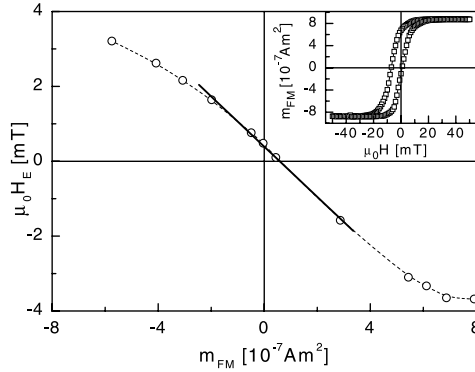


Fig. 1.  $H_e$  vs.  $m_{\text{FM}}$  dependence of the  $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2(110)/\text{Fe}14\text{nm}/\text{Ag}35\text{nm}$  heterostructure (circles). The data are obtained from the shifts of the magnetic hysteresis curves after cooling the system to  $T = 10\text{ K}$  in an applied field which sets the target value  $m_{\text{FM}}$ . The solid line shows a linear fit in accordance with proportionality (3). The inset shows a typical hysteresis after freezing in the saturated FM state.

obtained with  $m_{\text{FM}} = m_s$  where the exchange bias effect shifts the curve along the field line. Assuming that the FM interface moment  $S_{\text{FM}}$  is proportional to the net magnetic moment of the Fe layer during the freezing process, the simple MB formula predicts a linear  $H_e$  vs.  $m_{\text{FM}}$  dependence. In a first approximation the proportionality holds, but closer inspection shows that the data do not cross the origin of the coordinate system. Rather a small shift towards positive  $H_e$  values remains at  $m_{\text{FM}} = 0$ . Moreover, on approaching  $m_{\text{FM}} = \pm m_s$ ,  $H_e$  deviates from its linear  $m_{\text{FM}}$  dependence. The latter behavior indicates that the AF interface moment  $S_{\text{AF}}$  depends on both the freezing and the exchange field, which arises from the AF/FM interaction at the interface. Hence, its influence increases with increasing  $m_{\text{FM}}$ .

In the vicinity of  $m_{\text{FM}} = 0$ , however, it is reasonable to assume  $S_{\text{AF}} \approx \text{const}$ . As will be shown below, the shift of the  $H_e$  vs.  $m_{\text{FM}}$  curve agrees with the MB model, when generalizing the approach to a non-uniformly magnetized ferromagnet. In accordance with Ref. [12],  $\pm m_s$  yields  $\mp H_e$ . Hence, it is reasonable to start from the ansatz  $H_e = |H_e^+|a - |H_e^-|(1-a)$ . Here  $a$  is the relative portion of the total area of the Fe layer where the local magnetization is negative and, hence, the local exchange bias field is given by  $H_e^+ > 0$ , while  $(1-a)$  is the remaining part, where the magnetization is positive and  $H_e^- < 0$ . The shift of the total measured hysteresis is given by the sum of the local contributions weighted with respect to the relative areas. In accordance with the conventional MB formula,  $H_e^+$  and  $H_e^-$  are controlled by the local interface moments. It yields

$$H_e \propto |S_{\text{FM}}^+ S_{\text{AF}}^+|a - |S_{\text{FM}}^- S_{\text{AF}}^-|(1-a). \quad (1)$$

It is reasonable to assume that the magnitude of the FM interface moment per unit area does not depend on the

sign of the local magnetization,  $|S_{\text{FM}}^-| = |S_{\text{FM}}^+| = |S_{\text{FM}}|$ . However, the AF interface moment per unit area may depend on the orientation of the local FM interface moment  $|S_{\text{AF}}^-| = |S_{\text{AF}}^+| - \delta S_{\text{AF}}$ . A microscopic justification of a finite deviation,  $\delta S_{\text{AF}} \neq 0$ , is given below. Substitution of  $|S_{\text{AF}}^-|$  into Eq. (1) yields

$$H_e \propto |S_{\text{FM}}|((2|S_{\text{AF}}^+| - \delta S_{\text{AF}})a - |S_{\text{AF}}^+| + \delta S_{\text{AF}}). \quad (2)$$

The total magnetic moment of the Fe layer is given by  $m_{\text{FM}} = -m_s a + m_s(1-a)$ , the sum of domain contributions with positive and negative saturation magnetization. Hence, the normalized area can be expressed according to  $a = (1 - m_{\text{FM}}/m_s)/2$ . Substitution of  $a$  into Eq. (2) yields the explicit  $m_{\text{FM}}$  dependence of  $H_e$ . It reads

$$H_e \propto |S_{\text{FM}}| \left( \frac{-(2|S_{\text{AF}}^+| - \delta S_{\text{AF}})m_{\text{FM}}}{2m_s} + \frac{\delta S_{\text{AF}}}{2} \right). \quad (3)$$

Obviously, Eq. (3) describes the observed shift of  $H_e$  vs.  $m_{\text{FM}}$  (Fig. 1) in the case  $\delta S_{\text{AF}} \neq 0$ .

Whenever the AF bulk breaks into a domain state on cooling in a freezing field to below  $T_N$ , the magnitude and orientation of the AF interface moments are controlled by the competition between the exchange interaction with the adjacent FM layer and the adaptation of the interface spin configuration to the underlying AF domain structure. Only in the case of very strong exchange interaction at the interface, the ferromagnet will completely control the orientation of the AF interface moment so that  $S_{\text{AF}}^+ = -S_{\text{AF}}^-$ . However, in the case of a ‘strong’ antiferromagnet like  $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2$  a compromise between complete interface and bulk adaptation, respectively, has to be found. Hence,  $\delta S_{\text{AF}} \neq 0$  has to be expected in the case of AF domain states, which do not match perfectly with the FM ones.

It is well known, that a diluted AF in a field breaks into a random field domain state which carries a net magnetic moment. It gives rise to an AF interface moment and thus to exchange bias in the case of compensated AF surfaces [9]. However, comparison between the  $\text{Fe}_{1-x}\text{Zn}_x\text{F}_2(110)/14\text{ nm Fe}$  heterostructures with  $x = 0$  [3] and  $x = 0.4$  (this paper), reveals exchange bias of the same amount in the case of the compensated AF surface for both the undiluted [3] and the diluted heterosystem. In that case, the existence of an AF interface moment is usually attributed to crystal imperfections. Alternatively, we propose here an origin due to piezomagnetism [13,14] which was recently observed to cause vertical shift of the hysteresis curves of  $\text{FeF}_2(110)/\text{Fe}$  [3]. The same situation holds in our diluted heterostructure  $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2(110)/\text{Fe}5\text{ nm}/\text{Ag}35\text{ nm}$ , where for the first time the onset of a piezomagnetic moment on cooling to below  $T_N$  is determined. Fig. 2 shows the  $m$  vs.  $T$  data measured by SQUID-magnetometry with (squares) and without

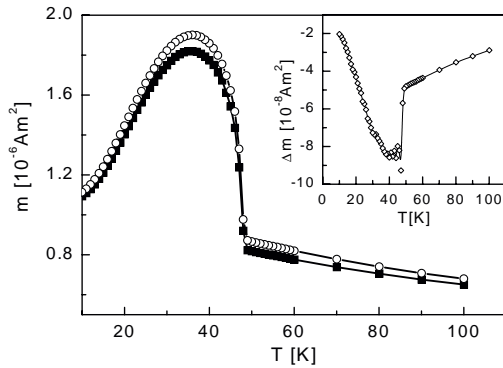


Fig. 2.  $m$  vs.  $T$  data measured with (squares) and without (circles) external shear stress  $\sigma_{xy} > 0$  applied along the  $[110]$  direction of the antiferromagnet. The inset exhibits the difference (diamonds) between both sets of data.

(circles) external shear stress  $\sigma_{xy} > 0$  applied along the  $[110]$  direction. The latter one modifies the piezomagnetic moment  $m_z^p = \lambda \sigma_{xy} l_z / |l|$ , by changing the natural stress distribution  $\sigma_{xy}(z)$ . Under an applied freezing field, the evolving piezomagnetic moment,  $m_z^p$ , will minimize its Zeeman energy. Hence, a built-in shear stress distribution  $\sigma_{xy}(z)$  with changing signs gives rise to changing signs of the AF vector  $\underline{l}$  and its  $z$  component  $l_z$ . Obviously, piezomagnetism creates an AF state which carries a magnetic moment and breaks into domains. Its interface contribution affects the exchange bias field in accordance with the MB approach. In addition, the domain formation will give rise to  $\delta S_{AF} \neq 0$ .

Fig. 3 shows the magnetic hysteresis after cooling from  $T = 100$  to 10 K in a freezing field of  $\mu_0 H = 5$  mT with (squares) and without (circles) external shear stress  $\sigma_{xy}$ . The  $[110]$  oriented stress originates from two copper plates which apply pressure on the top and bottom surfaces of the heterostructure. Copper wires which shrink on cooling connect the upper and lower plate and generate shear stress which reduces the piezomagnetic moment (see inset of Fig. 2). It indicates that the built-in stress has a negative sign on the average. Hence, in accordance with the MB model, the magnitude of  $H_e$  decreases from  $\mu_0 H_e = 25.3$  (Fig. 3 circles) to  $\mu_0 H_e = 23.1$  mT (Fig. 3 squares) on applying external shear stress. This behavior evidences the influence of the piezomagnetism on the exchange bias.

In conclusion we have shown that in the compensated  $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2(110)/\text{Fe}/\text{Ag}$  heterostructures the exchange bias field is controlled by the magnetic moment of the FM layer while the strength of the freezing field has minor influence. This is in contrast with, e.g., the strong freezing field dependence observed in the uncompensated  $\text{FeF}_2(001)/\text{CoPt}$  system, where the Zeeman energy of the interface moment competes with an AF interface exchange interaction. The latter one gives rise

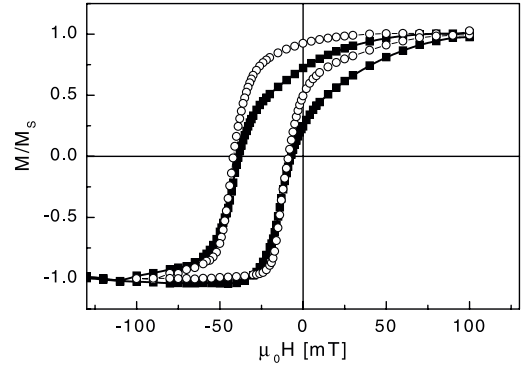


Fig. 3. Magnetic hysteresis after saturation of the FM layer at  $T = 100$  K and subsequent cooling in  $\mu_0 H = 5$  mT to  $T = 10$  K. Circles indicate the hysteresis under natural shear stress. Squares exhibit the hysteresis on applying external shear stress  $\sigma_{xy} > 0$ .

to a strong freezing field dependence of  $H_e$  above the saturation of the FM layer [5]. Details of the  $H_e$  vs.  $m_{\text{FM}}$  dependence are explained within a generalized MB model which takes into account both FM and AF domain formation. In addition to the established random field mechanism, we point out the impact of piezomagnetism on the exchange bias effect. Piezomagnetism is well known from antiferromagnets with rutile structure [13–15] and in the case of the  $\text{Fe}_{0.6}\text{Zn}_{0.4}\text{F}_2(110)/\text{Fe}/\text{Ag}$  heterostructure clearly evidenced from the steep increase of the magnetization on cooling to below  $T_N$ . However, its contribution to the AF interface moment is not quantitatively determined so far. Hence, presently it remains an open question whether piezomagnetism alone can be the origin of exchange bias. It is, however, worthwhile to take into account this mechanism which requires no impurities or structural defects in order to give a further possible explanation of the exchange bias in heterostructures with compensated both diluted and pure AF pinning layers which fulfill the necessary symmetry conditions [15].

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